Assimilation of sediments embedded in the oceanic arc crust: myth or reality?

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ABSTRACT

Arc magmas are commonly assumed to form by melting of sub-arc mantle that has been variably enriched by a component from the subducted slab. Although most magmas that reach the surface are not primitive, the impact of assimilation of the arc crust is often ignored with the consequence that trace element and isotopic compositions are commonly attributed only to varying contributions from different components present in the mantle. This jeopardises the integrity of mass balance recycling calculations. Here we use Sr and O isotope data in minerals from a suite of volcanic rocks from St Lucia, Lesser Antilles arc, to show that assimilation of oceanic arc basement can be significant. Analysis of $^{87}Sr/^{86}Sr$ in single plagioclase phenocrysts from four Soufrière Volcanic Complex (SVC; St Lucia) hand samples with similar composition ($^{87}Sr/^{86}Sr$ = 0.7089-0.7091) reveals crystal isotopic heterogeneity among hand samples ranging from 0.7083 to 0.7094 with up to 0.0008 difference within a single hand sample. $\delta^{18}O$ measurements in the SVC crystals show extreme variation beyond the mantle range with +7.5 to +11.1‰ for plagioclase (n=19), +10.6 to +11.8‰ for quartz (n=10), +9.4 to +9.8‰ for amphibole (n=2) and +9 to+9.5‰ for pyroxene (n=3) while older lavas (Pre-Soufriere Volcanic complex), with less radiogenic whole rock Sr composition ($^{87}Sr/^{86}Sr$ = 0.7041-0.7062) display values closer to mantle range: +6.4 to +7.9‰ for plagioclase (n=4) and +6 to +6.8‰ for pyroxene (n=5). We argue that the $^{87}Sr/^{86}Sr$ isotope disequilibrium and extreme $\delta^{18}O$ values provide compelling evidence for assimilation of material located within the arc crust. Positive correlations between mineral $\delta^{18}O$ and whole rock $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$ and $^{206,207,208}Pb/^{204}Pb$ shows that assimilation seems to be responsible not only for the isotopic heterogeneity observed in St Lucia but also in the whole Lesser Antilles since St Lucia

encompasses almost the whole-arc range of isotopic compositions. This highlights the need for detailed mineral-scale investigation of oceanic arc suites to quantify assimilation that could otherwise lead to misinterpretation of source composition and subduction processes.

1.Introduction

A key question in oceanic arc geochemistry concerns the relative contributions of subducting slab and intra-crustal material in the chemical and isotopic characteristics of lavas. Oceanic arc magmas are generated by partial melting of the mantle wedge modified by H_2O-rich fluids and melts from the subducting slab (Tatsumi and Eggins, 1995) and subsequently ascend through the arc crust before eruption. Lavas erupted at oceanic arcs rarely have major element compositions in equilibrium with mantle peridotite (Annen et al, 2006) which suggests that the magmas experienced differentiation during storage within or at the base of the arc crust. Depending on the nature of the arc basement, such magmas may interact with igneous or metasedimentary wall rocks during differentiation. Distinguishing between sediment addition to the mantle wedge and assimilation of metasediments located in the arc crust is not straightforward on the basis of whole rock compositions alone– the effects of mixing sediment into the mantle wedge or with basaltic melts in the crust are similar in most radiogenic isotope-isotope spaces.

In order to investigate the role of crustal assimilation in oceanic arcs we selected a suite of rocks from the Lesser Antilles arc where, despite the absence of continental basement, whole rock isotope ratios of arc lavas can be very "continental". As a result, the case has long been made both for crustal assimilation (e.g. Davidson, 1987; Davidson and Harmon, 1989; Smith et al., 1996; Thirlwall et al., 1996; Thirlwall and Graham, 1984; Van Soest et al., 2002), and incorporation of sediment or sediment melt into the mantle wedge (e.g. Carpentier et al., 2008; 2009; Labanieh et al., 2010; 2012; White and Dupré, 1986). Our suite of samples, from the island of St Lucia, encompasses almost the entire range of whole rock isotopic compositions observed in the Lesser Antilles, ranging from values close to

typical intra-oceanic arc rocks to those resembling continental crust (e.g. $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$ ranging from 0.70411-0.70906, 0.51210-0.51298 and 19.291-19.797 respectively). $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ analyses of individual minerals separated from lavas with extreme crust-like whole rock isotopic composition from the Soufriere Volcanic Complex (SVC), demonstrate the importance of open system behaviour on oceanic arc magmatism and challenge our understanding of magma differentiation in oceanic arcs.

2.Geological background

The Lesser Antilles arc (Fig. 1) formed as a result of subduction of the North American Plate under the Caribbean Plate. Its lavas have typical oceanic arc compositions in the northern section but unusually heterogeneous isotopic signatures in the central and southern part of the arc where both typical intra-oceanic arc and very continental crust-like signatures are observed (e.g. Macdonald et al., 2000; Fig.2). Two main processes have been proposed to explain these extreme compositions: (1) incorporation of sediment into the mantle source or (2) significant assimilation of sediment-rich arc crust. While a high sediment input to the source could be explained by the presence of abundant sediment in the southern Antilles Trench, due to discharge from the Orinoco and Amazon rivers (e.g. Carpentier et al., 2008; 2009), assimilation of sediment in the arc crust is also possible because the central-southern Lesser Antilles arc is thought to have developed above the thick forearc basin of the (now extinct) Aves Ridge Arc, splitting it into the Grenada and the Tobago basins (Fig. 1; Aitken et al., 2011). Sediments entering the subduction zone are chemically and isotopically well constrained by analyses from DSDP Sites 144 and 543 (Fig.1; Carpentier et al., 2008; 2009; White and Dupré, 1986). In contrast, the nature of the basement of the Southern Lesser Antilles arc is still poorly known. While the detrital

sediments from the South American craton, which dominate the sequences on both the subducting and the overriding plates, were often targeted in the past to explain the high $^{87}Sr/^{86}Sr$ and low $^{143}Nd/^{144}Nd$ ratios of the lavas, they fail to explain their very radiogenic Pb signatures. Recently, analyses of Mesozoic black shales sampled at DSDP site 144 showed that these sediments have sufficiently radiogenic Pb to account for the compositions of the lavas from the southern arc (Carpentier et al., 2009). Because such radiogenic Pb has not yet been reported from the sediments of the Grenada and Tobago basins, sediment addition to the source alone has been suggested to explain the whole range of isotopic data in the southern arc lavas (Carpentier et al., 2008; 2009; Labanieh et al., 2010). However, it cannot be ruled out that such sediments exist within the basement of the southern arc since biogenic rich limestones have been reported in the overriding plate, in the Late Cretaceous to Oligocene sequences of the Carupano Basin (Ysaccis, 1997).

St Lucia is an island in the central-southern part of the arc (Fig.1). The lavas from this single island encompass almost the whole range of isotopic variation observed in the arc making it the perfect location to study the influence of crustal components (Fig.2). The more continental crust-like compositions are found in andesites (3Ma to ca. 250 ka) and dacites (100 ka to present) of the Soufriere Volcanic Complex (SVC), while more mantle-like compositions are observed in the more mafic Pre-SVC lavas dominated by basalt, basaltic andesite and andesite (18 Ma to 1.1 Ma; Briden et al., 1979; De Kerneison et al., 1983; Lindsay et al., 2013; Samper et al., 2008; Schmidt et al., 2010).

3.Methods

3.1.Whole rock Sr, Nd and Pb isotopes

Except for 7 samples in which Sr and Nd isotope ratios were measured at the Geochemical Analysis Unit at Macquarie University (Aus), whole rock powders were analysed at the Arthur Holmes Isotope Geology Laboratory (AHIGL) which is part of Durham Geochemistry Centre (DGC) at Durham University (UK) At both institutes, 0.1g of sample powder was dissolved in Teflon distilled 29M HF and 16M HNO_3.

At Durham, Pb and Sr were separated from the sample solution using Sr-spec resin columns. Nd was collected from the same Sr-spec column before being passed through a cation column where it was collected as part of a total REE cut after elution of Hf, Rb and Ba. Samples were analysed for their $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$ and $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ compositions by plasma ionisation multicollector mass spectrometry (PIMMS) using a Thermo Scientific Neptune instrument. During the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ analytical sessions, instrument performance was monitored by analysis of NBS987 Sr standard and an in house J&M Nd standards and within-run instrument mass fractionation was corrected using an exponential law and the normalising value of $^{88}Sr/^{86}Sr = 8.375209$ and $^{146}Nd/^{145}Nd = 2.079143$ respectively (equivalent to $^{86}Sr/^{88}Sr = 0.1194$ and $^{146}Nd/^{144}Nd = 0.7219$). Since Nd was analysed as part of a total REE cut, the data requires an algebraic correction for Sm interference on Nd based on the approach of Nowell and Parrish (2001). The accuracy of this correction was monitored by analysis of Sm-doped J&M, with a Sm/Nd ratio of ~0.25.The average $^{87}Sr/^{86}Sr$ for NBS987 was 0.710272 ± 0.000020 (2sd; n=14) and the average $^{143}Nd/^{144}Nd$ for both pure and Sm-doped J&M was 0.511105 ± 0.00002 (2sd; n=15). $^{87}Sr/^{86}Sr$ ratios are reported relative to NBS987 standard value of 0.71024 (Thirlwall, 1991) and $^{143}Nd/^{144}Nd$ is reported relative to a J&M standard value of 0.511110 which is equivalent to a La Jolla value of 0.511862 (Royse et al., 1998). The

$^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios of the two international rock standards BHVO-1 and BIR-1 were 0.703463 and 0.512989 and 0.703115 and 0.513073 respectively. The accuracy of the Sm correction on the $^{143}Nd/^{144}Nd$ ratio is illustrated by the analysis of BHVO-1, which after Sm correction is identical to the ratio of 0.512986 ± 0.000009 (2sd; n=19) that Weiss et al. (2005) obtained by thermal ionisation mass spectrometry (TIMS).

Following chemistry, the Pb fractions were taken up in 1 ml of 3% HNO_3. The Pb concentration of the aliquots was analysed before isotopic measurements, in order to calculate the appropriate amount of Tl spike to add to obtain a Pb/Tl ratio of ~12. This minimizes the tail from ^{205}Tl onto ^{204}Pb and from ^{206}Pb onto ^{204}Tl. During isotopic measurements, the samples were introduced into the Neptune using an ESI PFA50 nebulizer and a cyclonic spray-chamber. The normal H skimmer cone was used. Sensitivity for Pb on the Neptune using such setup is typically around 100 V total Pb ppm^{-1} at an uptake rate of 90 µm min^{-1}. Pb mass bias was corrected externally using the $^{205}Tl/^{203}Tl$ ratio of the spike and an exponential law. The $^{205}Tl/^{203}Tl$ used for correction was determined for each analytical session by minimizing the difference in offset between the session average Pb ratios and the Galer (1997) triple spike Pb isotope values. The Tl isotope ratio was calculated to yield the best fit to all the Pb isotope ratios of Galer (1997) simultaneously. During the analytical sessions, the NBS981 standard solution was analysed regularly (n=16). The average ratios were: $^{206}Pb/^{204}Pb = 16.941 \pm 0.0024$ (2 sd), $^{207}Pb/^{204}Pb = 15.497 \pm 0.0012$ (2 sd), $^{208}Pb/^{204}Pb = 36.715 \pm 0.0039$ (2 sd). Furthermore, a total procedural BHVO-1 was analysed. The $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ ratios obtained were 18.687, 15.568 and 38.342, respectively which is in good agreement with the GEOREM

154 accepted value ($^{206}Pb/^{204}Pb$ = 18.692 ±0.008 (2 sd); $^{207}Pb/^{204}Pb$ = 15.572 ± 0.006 (2 sd);
155 $^{208}Pb/^{204}Pb$ = 38.355 ± 0.022 (2 sd)).

156

157 At Macquarie, Sr was separated from the sample solution using Biorad AG50W-X8 resin
158 and Nd was collected after separation from Ba and LREE using Eichrom® Ln.spec resin
159 columns following the method of Pin et al. (1997). $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios were
160 measured by TIMS using a Thermo-Fisher Triton instrument. During the analytical session,
161 instrument performance was monitored by analyses of Sr standards NBS987 and Nd
162 standard JMC321 and mass bias was corrected as in Durham. The average $^{87}Sr/^{86}Sr$ for
163 NBS987 was 0.710220 ± 0.000022 (2sd, n=4) and the average $^{143}Nd/^{144}Nd$ for JMC321
164 was 0.511123 ± 0.000006 (2sd; n=3). $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ were not reported relative
165 to a literature value. During the course of this study, analyses of processed international
166 BHVO-2 standard yielded average $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios of was 0.703469 ±
167 0.000012 (2sd; n=4) and 0.512976 ± 0.000012 (2sd; n=4), respectively.

168 *3.2. $^{87}Sr/^{86}Sr$ in plagioclase*

169 Four of the most continental-like lavas from St Lucia (SVC), ranging from andesitic to
170 dacitic in composition and with very similar whole rock radiogenic isotope signatures, were
171 chosen for in-depth isotopic study. Sr isotope ratios were determined for five single
172 plagioclase crystals from each lava. The crystals were carefully hand-picked from lightly
173 crushed hand specimens using a binocular microscope. Grains were selected for being free
174 of both inclusions and adhering glass. Each grain, containing at least 60 ng Sr, was
175 individually digested in Romil Upa grade HNO_3 and HF. Sr was separated using Sr-spec

176 resin columns and $^{87}Sr/^{86}Sr$ was measured by TIMS using the Thermo Fisher Triton at
177 Durham University. During the TIMS analyses, the average $^{87}Sr/^{86}Sr$ ratio for NBS987
178 obtained on 12ng of Sr was 0.710243 ± 0.000009 (2sd, n=3). This is in excellent agreement
179 with the value reported by Thirlwall (1991) of 0.710248 ± 0.000023 (2sd, n=427). Two
180 total procedure blanks contained 18 and 32 pg of Sr which represent 0.03 and 0.05% of the
181 lowest sample Sr concentration analysed and are therefore negligible.

182 *3.3. δ18O in mineral separates*

183 Samples ranging from typical oceanic arc lavas to the most continental-like composition
184 were selected for individual mineral oxygen isotope extraction by laser-fluorination.
185 Minerals separates were hand-picked from lightly crushed hand specimens under a
186 binocular microscope, avoiding any inclusions or glass adhering to the grains. Oxygen was
187 extracted from 0.9-3.7 mg of separate using a total laser fluorination system based on the
188 method of Sharp (1990), at the Scottish Universities Environmental Research Centre
189 (SUERC). All fluorinations resulted in 100% release of O_2 from mineral lattice. This
190 oxygen was converted to CO_2 and analyzed on a VG Optima mass spectrometer. Oxygen
191 isotope ($\delta^{18}O$) values are reported as per mil (‰) deviations relative to Vienna Standard
192 Mean Ocean Water (V-SMOW). Samples were analysed during two periods. During the
193 first period (March 2012) 31 unknowns were analysed along with the international and in-
194 house standards UWG2 and SES and GP147. The average $\delta^{18}O$ values obtained for UWG2
195 (garnet, +5.7‰ ± 0.2 (2sd), n = 11), SES (quartz, +10.4‰ ± 0.6 (2sd), n = 10) and GP147
196 (garnet, +7.2‰ ± 0.4 (2sd), n = 3) are in very good agreement with their accepted values
197 of +5.8‰ (Valley et al., 1995), +10.2‰ and +7.2‰ (Mattey and Macpherson, 1996).
198 respectively. The internal SES standard has been run many hundreds of time over the past

20 years in SUERC, and is well calibrated against UWG2, NBS 28 and NBS 30, as well as GP147. Twelve unknowns were analysed during the second period of study (November 2012) when the average $\delta^{18}O$ values for UWG2, SES and GP147 were +5.9‰ ± 0.2 (2 sd, n = 2), +10.2‰ ± 0.3 (2 sd, n = 6), +7.15‰ ± 0.4(2 sd, n = 2) respectively.

4.Results

Whole rock Sr, Nd and Pb isotope ratios are given in Table 1. Plagioclase Sr isotopes and mineral $\delta^{18}O$ data are presented in Tables 2 and 3 respectively.

Whole rock $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios range between 0.70411 and 0.70622 and 0.51251 and 0.51298, respectively, in the Pre-SVC samples and from 0.70754 to 0.70906 and 0.51210 to 0.51226, respectively, in the SVC samples (Fig. 2a). Whole rock $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios were analysed in selected samples with the highest and the lowest $^{87}Sr/^{86}Sr$ isotope ratios, to verify coupling with other isotopic systems. Pb isotope ratios vary from 19.291 to 19.341 for $^{206}Pb/^{204}Pb$, from 15.747 to 15.748 for $^{207}Pb/^{204}Pb$ (Fig. 2b) and from 38.96 to 39.11 for $^{208}Pb/^{204}Pb$ in the Pre-SVC lavas analysed. In the SVC lavas, ratios vary from 19.721 to 19.797 for $^{206}Pb/^{204}Pb$, from 15.826 to 15.846 for $^{207}Pb/^{204}Pb$ and from 39.438-39.528 for $^{208}Pb/^{204}Pb$. Plagioclase crystals from the SVC have heterogeneous $^{87}Sr/^{86}Sr$ ratios (Fig. 3) ranging between 0.7083 and 0.7094 with variations of up to 0.00083 among crystals from a single lava (SL-JL-51). In the Pre-SVC, plagioclase and pyroxene $\delta^{18}O$ vary from + 6.4 to +7.9‰ and from +6 to +6.8‰ respectively (Fig. 4). In the SVC, mineral $\delta^{18}O$ are higher and vary from +7.5 to +10.9‰ for plagioclase, from +10.6 to +11.8‰ for quartz, from +9.4 to +9.8‰ for amphibole and +9 to +9.5‰ for pyroxene.

221 **5.Discussion**

222 *5.1.Intra-crustal magma contamination*

223 The unusually radiogenic composition of some lavas from the Lesser Antilles arc has been
224 explored by many authors using whole rock samples, and explained through models
225 invoking either sediment incorporation into the source (e.g. Labanieh et al., 2010) or
226 assimilation during fractional crystallisation (AFC; e.g. Davidson, 1987). Discriminating
227 between these two processes using only whole rock isotopes is difficult mainly due to
228 uncertainties in determining the precise composition of the endmembers. Correlations
229 between whole rock isotopes and indexes of differentiation such as SiO_2 or MgO are
230 however useful in detecting isotopic changes during differentiation and have been
231 employed by several authors to argue for AFC in Grenada (Thirlwall et al., 1996), the
232 Grenadine Islands (Smith et al., 1996) and Martinique (Davidson, 1987; Davidson and
233 Wilson, 2011). In St Lucia, two distinct trends are observed (Fig. 2c): the first one,
234 comprising only some Pre-SVC samples (Pre-SVC1), shows an absence of covariation
235 between radiogenic isotopes and SiO_2 or MgO while the second trend, comprising the
236 remaining Pre-SVC (Pre-SVC2) and the SVC lavas, displays progressive increase in Sr
237 and decrease in Nd isotope ratios with increasing SiO_2 and decreasing MgO. Pb isotope
238 ratios were not analysed for all St Lucia lavas but variations in the Pre-SVC1 and SVC
239 lavas are consistent with those defined by Sr and Nd isotopes (Fig. 2d). While the first
240 ("vertical") trend can be explained by simple differentiation of a mafic mantle-derived
241 magma, the second trend requires a different/additional process. If this latter trend were to
242 reflect a source process, this would require either (1) the production of silicic SVC magmas
243 in the mantle or (2) their derivation by differentiation of more mafic melts from a mantle

source where a common factor controls both the amount of sediment melt/sediment derived fluids added to the source and the future extent of magma differentiation. The first hypothesis cannot be reconciled with the low MgO content of silicic SVC lavas compositions (MgO = 1.1-2.7 wt.%) since andesitic to dacitic magma generated either by mantle melting or by reaction between ascending slab-derived silicic melts and mantle peridotite typically have elevated MgO contents (Grove et al., 2003; Yogodzinsky and Kelemen, 1998). The second hypothesis, involving a coincidental process, is equally very hard to conceive. It is, however, directly testable. Since magmatic differentiation has negligible effect on radiogenic isotopic ratios, the amount of sediment incorporated into the magma during its genesis in the mantle should be reflected in the $^{87}Sr/^{86}Sr$ of all its components. Therefore, once separated from its source, a magma that has remained a closed system should demonstrate negligible $^{87}Sr/^{86}Sr$ variation between the melt and all phenocrysts. Furthermore, the Sr isotope ratio of the phenocrysts would also be observed for the whole rock. Conversely, isotopic disequilibrium between different phases in a rock or between different crystals of the same phase would require that open system behaviour occurred. Therefore, the variation in $^{87}Sr/^{86}Sr$ ratios within single samples is a hallmark for open system behaviour (Davidson et al., 2007).

Despite their similar whole rock Sr isotopic ratios, the $^{87}Sr/^{86}Sr$ ratios of crystals from the four SVC samples vary substantially amongst and within hand specimens (Table 2; Fig.3). Such disequilibrium cannot be explained by incorporation of sediment in the source, and must instead be accounted for by an open system process, such as crustal assimilation during crystallisation.

Most of the plagioclase crystals analysed have lower $^{87}Sr/^{86}Sr$ than their respective whole rock values. This indicates that at least part of the volume of each of those plagioclase crystals grew in a magma with less radiogenic Sr than now represented by the matrix. The zoned nature of the crystals requires an igneous origin but does not allow discrimination between phenocrysts which grew from a magma changing in $^{87}Sr/^{86}Sr$ composition and xenocrysts which were remobilized during re-melting of plutonic rocks. The latter option was preferred by Schmitt et al. (2010) to explain variation in core to rim U-Th zircon ages obtained in the SVC lavas.

5.2. Effect of contamination on whole rock compositions

In order to confirm that crustal assimilation is largely responsible for the radiogenic whole rock isotopic signatures observed in St Lucia, the mineral $\delta^{18}O$ study was performed. Although the $\delta^{18}O$ of crustal-derived material (> +10‰) is much higher than the mantle range (+ 5.5‰ ± 0.2), 80% of the $\delta^{18}O$ values of any given phase in oceanic arc lavas fall within ± 0.2‰ of the average value for that phase in upper mantle peridotites and MORBs (Eiler et al., 2000; Mattey et al., 1994). Because the concentration of oxygen in the mantle and the crust is similar, a large input of crustal derived fluids and/or sediments would be necessary to modify the mantle signature (James, 1981). Thus, O isotopes provide a powerful tool to discriminate between addition of sediments to the mantle source from assimilation of sediments in the arc crust, the latter being a much more efficient way of modifying the mantle-derived $\delta^{18}O$ value of the magma (e.g. Macpherson et al., 1998). Previous oxygen isotopic analyses performed on olivine and pyroxene phenocrysts from the Lesser Antilles showed the existence of values greater than the mantle range and that

these correlate with whole rock radiogenic isotopes, suggesting crustal assimilation (Smith et al., 1996; Thirlwall et al., 1996; Van Soest et al., 2002). However, although higher, the excess relative to the range of mantle $\delta^{18}O$ values was small, with only up to + 1‰ excess for olivine and up to + 0.6‰ for pyroxene with the exception of one pyroxene, which was + 1.6‰ and two quartz crystals which were + 3.83‰ and + 4.37‰ higher than the mantle range.

In this study, 93% of the 44 mineral analyses lie outside their respective mantle ranges and the SVC displays extreme $\delta^{18}O$ values never observed before in oceanic arcs lacking known continental basement (Fig.4). Moreover, a strong positive correlation is observed between the mineral $\delta^{18}O$ and the whole rock radiogenic isotopes (Fig. 5) with the highest $\delta^{18}O$ values found in SVC lavas with very radiogenic whole rock $^{87}Sr/^{86}Sr$ and $^{206,207,208}Pb/^{204}Pb$ ratios (and unradiogenic $^{143}Nd/^{144}Nd$ ratios). Although the SVC products have more differentiated compositions than the pre-SVC samples, the maximum effect of closed system differentiation on $\delta^{18}O$ is typically very small: +0.3 to +0.4‰ (Bindeman et al., 2004; Macpherson and Mattey, 1998). Thus, even after correction for this effect, the SVC mineral $\delta^{18}O$ range lies well beyond the mantle range. The mean anorthite (An) content of SVC plagioclase is slightly lower than that of the pre-SVC crystals. However, since the difference is small (mean An of 68% (ranging from 65% to 71%) and of 59% (ranging from 54% to68%) respectively for SVC andesite and dacite vs. mean An of 80% (70% - 90%) for Pre-SVC basaltic andesite and basalt) the related increase in $\delta^{18}O$ in the SVC plagioclase is thought to be negligible. This is confirmed by the fact that phenocrysts from SVC lavas with different SiO_2 (high-silica andesites and dacites: 60.5 – 67.5 wt. %) possess similar $\delta^{18}O$ values. SVC plagioclase-quartz pairs (from the same hand sample)

311 have $\Delta_{plag\text{-}qtz}$ (where $\Delta_{i\text{-}j} = \delta^{18}O_i - \delta^{18}O_j$) between -0.1 to -1.21 ‰ (n = 8; with n=5 higher
312 than -0.7‰) except for one pair where $\Delta_{plag\text{-}qtz}$ = -3.9 ‰. Andesitic-dacitic temperatures
313 (T= 800-1000°C) typically produce $\Delta_{plag\text{-}qtz}$ between -0.84 and -1.45 ‰ (e.g. using the
314 Chiba et al. (1989) fractionation coefficients and considering a large range of anorthite
315 content between An_{40} and An_{70}). Therefore, most $\Delta_{plag\text{-}qtz}$ are higher than equilibrium. SVC
316 plagioclase-pyroxene pairs display fractionation of $\Delta_{plag\text{-}py}$ between +0.60 and +0.95 ‰
317 (n=3) which are similar to that expected from equilibrium fractionation at andesitic-dacitic
318 temperatures ($\Delta_{plag\text{-}py}$ = +0.66 to +1.21 ‰ at T° = 800-1000 and $An_{40\text{-}70}$). Therefore, both
319 pyroxene and plagioclase are likely to have crystallized in equilibrium from a similar
320 magma. The lowest $\delta^{18}O$ values are observed in Pre-SVC1 lavas, which have low whole
321 rock $^{87}Sr/^{86}Sr$ and Pb isotope ratios (SL-83-25, 44). In these samples, $\delta^{18}O_{plag}$ values are
322 higher than the mantle range, whereas the $\delta^{18}O_{px}$ values overlap the upper end of the mantle
323 range. However, plagioclase-pyroxene pairs ($\Delta_{plag\text{-}py}$ from +0.3 to +0.6 ‰) are close to
324 oxygen isotopic equilibrium at basaltic to basaltic andesite temperatures ($\Delta_{Plag\text{-}py}$ = +0.35
325 ‰ to +0.51‰ at T° = 1100-1200°C and $An_{99\text{-}80}$ using Chiba et al.'s (1989) equation). Thus,
326 it is difficult, in these samples, to differentiate between small error on the mantle mineral
327 range of composition and the impact of small amounts of crustal assimilation during
328 plagioclase crystallization. Pre-SVC2 lavas, that have the highest whole rock $^{87}Sr/^{86}Sr$
329 ratios (SL-83-26; $^{87}Sr/^{86}Sr$ = 0.706219; basaltic andesite) of the Pre-SVC group, also
330 display higher $\delta^{18}O_{plag}$ than $\delta^{18}O_{px}$. However, $\Delta_{plag\text{-}py}$ varies between +0.5 ‰ to +1.5 ‰.
331 Such values are too high to be explained simply by equilibrium fractionation. We believe
332 that the high $\Delta_{plag\text{-}py}$ values result from the addition of a crustal component late in the
333 differentiation sequence when plagioclase formed a larger component of the crystallising

assemblage. This provides further support for an origin through crustal assimilation rather than sediment addition to the source. In this case, plagioclase provides a more sensitive record of crustal assimilation than the mafic phases. This may be important when targeting minerals for oxygen isotopic analysis in settings where more subtle changes in $\delta^{18}O$ are anticipated, due to smaller isotopic contrasts between magma and the crust into which it is emplaced.

A simple mixing model (Fig. 5) between a typical mantle composition and a sediment having the highest $\delta^{18}O$ reported (+ 35 ‰; Bindeman, 2008) shows that a minimum of 10-20 % of sediment would be required to be added to the mantle or a mantle derived magma in order to explain the SVC mineral $\delta^{18}O$ data. However, we know from DSDP 78A site 543 and DSDP 14 site 144 that sediments subducted at the Lesser Antilles trench comprise a mixture of pelagic clay, radiolarian clay, terrigeneous claystone siltstone and sandstones (30% carbonate on average; Carpentier et al., 2008). Thus, the bulk sediment would possess an average $\delta^{18}O$ closer to the composition of site 543 pelagic clays of +20‰ (Davidson, 1987). In this more realistic case, addition of 20-40% sediment would be required to generate the range of SVC $\delta^{18}O$ values and addition of 10-20% sediment could reproduce the Pre-SVC sample with the most elevated $\delta^{18}O_{plag}$ (+7.3 to +7.9‰ in SL-83-26; Fig. 5c). Introducing such large amounts of sediments to the mantle wedge should also modify the major element composition of the primitive magmas, such that increasing sediment incorporation (i.e. increasing mineral $\delta^{18}O$ and $^{87}Sr/^{86}Sr$) should correlate with an increase of SiO_2 (clay, sand) or CaO (carbonate) of the melt. As discussed in section 5.1., the SVC lavas cannot have been directly produced in the mantle since their MgO is too low. If the SVC lavas resulted from differentiation of a primary magma produced in the mantle by

357 mixing with substantial amounts of sediment melt, the resulting suite of lava would likely
358 have a higher SiO_2 or CaO content than a suite derived from Pre-SVC1,2 primitive magmas
359 at a given MgO content (different differentiation trends). However, SVC and Pre-SVC1,2
360 lavas share similar (although scattered) differentiation trend (Fig. 6), and therefore back
361 project toward a similar primitive magma. Hence, it seems unlikely that the SVC and Pre-
362 SVC lavas were derived from very different mantle sources. Instead, the single trend of
363 differentiation recorded by major elements in all St Lucia lavas is consistent with variable
364 assimilation of crustal material. This is because, during differentiation, major element
365 compositions are largely controlled by phase equilibria and, to a lesser extent, by the nature
366 of the sediment assimilated, the latter likely to be derived from the erosion of upper
367 continental crust with a similar composition to the felsic differentiates of the Pre-SVC lavas
368 (Fig. 6). Therefore, the strong correlation between $\delta^{18}O$ and whole rock Sr isotopes indicate
369 that assimilation is the major factor controlling the whole rock radiogenic isotope ratios
370 observed in the St Lucia lavas (Fig.7). For both plagioclase and pyroxene, the correlation
371 is slightly convex-up, suggesting that the magma had a slightly lower Sr content than the
372 material it assimilated (c.f. inset to Fig.5). More importantly, the plagioclase $\delta^{18}O$ values
373 are displaced further from the mantle field than is the case for pyroxene in Pre-SVC
374 samples, requiring a larger proportion of assimilant than required for the pyroxenes. Quartz
375 is only found in the SVC lavas where no correlation between $\delta^{18}O_{qz}$ and whole rock
376 $^{87}Sr/^{86}Sr$ is observed. The slight disequilibrium between quartz and plagioclase (low Δ_{plag-}
377 $_{qtz}$) indicates that quartz crystallized in a magma with slightly lower $\delta^{18}O$ than the
378 plagioclase.

379 *5.3. Implications for other Lesser Antilles volcanoes and other arcs*

The crystal $^{87}Sr/^{86}Sr$ data show that lavas with extreme whole rock compositions underwent assimilation of crustal material in an open system supporting previous work by Davidson (1987), Davidson and Harmon (1989) Smith et al. (1996), Thirwall et al. (1996), Thirlwall and Graham (1984) and Van Soest et al. (2002). The extreme $\delta^{18}O$ signature observed in lavas with the most continental Sr, Nd and Pb isotope ratios (SVC) confirms late assimilation of arc crust as the cause of these variations and shows that the amount of material assimilated is not trivial (>20%). More importantly, crustal assimilation did not only affect the andesites and dacites, but also substantially (10-20% assimilation) modified the composition of a basaltic andesite (Pre-SVC2) with similar SiO_2 and MgO contents to the most mafic lava of the island. Therefore, in St Lucia, the use of the most mafic lavas erupted on the island to constrain the source characteristics is compromised since some of them have clearly interacted with the crust. Similar problems may affect other Lesser Antilles islands and other oceanic arcs where basaltic andesites with MgO ~ 4 wt. %, or even more differentiated lavas, have been used studies to constrain the source composition. For example Labanieh et al. (2010) suggested that isotopic ratios in lavas from Martinique with up to ~70 wt. % SiO_2 (MgO ~ 0.23) faithfully reflect the source composition. Likewise, Carpentier et al. (2008) modeled all the published data for Lesser Antilles lavas by sediment addition to the source, without taking into account the degree of differentiation of the lavas.

Because St Lucia lavas encompass Sr, Nd and Pb isotopic range almost as great as the whole Lesser Antilles arc it follows that much of the isotopic heterogeneity observed in the rest of the arc could also reflect variable crustal assimilation. Sediment present in the Lesser Antilles arc crust is likely to originate from the closest continental mass: South America.

It has very "continental" $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$ and $^{206,207,208}Pb/^{204}Pb$ ratios (e.g. Carpentier et al., 2008). Sediments with such signatures were found on the edge of the north east part of South America (site 144 sediment; Carpentier et al.(2008, 2009)) and their presence on the subducting plate was used as a strong argument for incorporation of sediment in the mantle source as being the cause for the extreme isotopic compositions of the arc lavas. However, our data show that most of the arc lava isotopic variations are generated by contamination in the arc crust. Therefore, subduction of material resembling site 144 sediments is not *required* to explain the extreme Sr, Nd, Pb and O isotopic signatures of the Lesser Antilles arc lavas.

It is important to stress that this does not preclude incorporation of subducted sediment into the source of the lavas having an impact on the whole rock isotopic ratios. Indeed, mineral $\delta^{18}O$ compositions of the Pre-SVC1 lavas (SL-83-44; SL-83-25) are very close to mantle values which indicate that, if crustal assimilation affected these lavas, it would be in very low amounts. Yet, although their Sr and Nd isotope ratio overlap typical oceanic arc compositions, their Pb isotope ratios are slightly more radiogenic (Fig. 2b). This not only suggests the involvement of sediment in the source but also that these are slightly more abundant or more radiogenic than sediments present in the source of most "typical" oceanic arcs. However, Sr, Nd and Pb isotope compositions of the Pre-SVC1 lavas remain very close to typical oceanic composition spectrum, and we suggest that subducted material is not a major factor in causing large isotopic heterogeneity in Lesser Antilles arc lavas.

The extreme contamination observed in at least some of the Lesser Antilles lavas may reflect the unique presence of sediment in the arc crust that, in turn, reflects a specific geodynamic context (Aitken et al., 2011). However, similar assimilation of sediments and/or altered crust in lower amounts and/or different composition may occur in other oceanic arcs and has already been suggested to explain compositions observed in the offshore Taupo Volcanic zone in New Zealand (Macpherson et al., 1998). Eiler et al. (2000) estimated that although most oceanic arc minerals (olivine, plagioclase, glass, biotite) fall within the mantle range in $\delta^{18}O$, around 20% of the data are slightly higher by up to 0.43‰. These `out of mantle range` phases come from samples with higher whole rock Sr isotope ratios, which is also what we observe on St Lucia. Because the displacement from the mantle range is not substantial, these data have been interpreted to reflect sediment incorporation into the mantle wedge. However, all these higher $\delta^{18}O$ ratios were observed in mafic minerals (primarily olivine). At St Lucia, in the least contaminated rocks, plagioclase reveals assimilation more clearly than pyroxene, probably because it crystallised over a greater range of magmatic evolution, persisting to the most differentiated stages. Hence, comparing olivine and pyroxene $\delta^{18}O$ values with whole rock $^{87}Sr/^{86}Sr$ may be a less effective means to detect small amounts of assimilation than the use of plagioclase $\delta^{18}O$ data.

6. Summary and conclusions

Our new single plagioclase Sr isotopic data and mineral $\delta^{18}O$ data show that crustal assimilation was important in magma evolution at St Lucia. Correlations of mineral $\delta^{18}O$ values with whole rock Sr isotopic composition shows that assimilation controls most of the Sr, Nd and Pb isotopic variation in St Lucia lavas. Isotopic variation at St. Lucia

447 replicates most of the range for the whole Lesser Antilles arc, therefore we suggest that up
448 to 20-40% assimilation could be responsible for the unusual diversity of isotopic
449 compositions observed along the arc. Assimilation of such large volumes of sediment
450 within the Lesser Antilles crust may be related to a specific geographic and geodynamic
451 setting. However, similar crustal assimilation could take place in other oceanic arcs where
452 a lack of geochemical contrast between assimilated sediments and the magma and/or the
453 lower amounts of sediments assimilated may make it more challenging to track.

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589 **Figure captions**

590 Figure 1: Bathymetric map of the Lesser Antilles arc showing the islands, the trench-
591 deformation front, the main ridges, the Grenada and Tobago basins and the locations of
592 Deep Sea Drilling Poject (DSDP) hole 543/543A and hole 144. Figure modified from Van
593 Soest et al. (2002).

594 Figure 2: Whole rock compositions of St Lucia lavas. (a) Sr and Nd and (b) Pb and Nd
595 isotopic diversity. St Lucia data show a similar range to most of the Lesser Antilles arc,
596 ranging between typical oceanic arc compositions and Atlantic sediments cored at the front
597 of the trench (DSDP sites 144 and 543; Carpentier et al., 2008, 2009). SVC = Soufriere
598 Volcanic Complex. Pre-SVC lavas with typical oceanic arc compositions, called Pre-
599 SVC1, are shown by open circles and lavas with the most radiogenic compositions, called
600 Pre-SVC2, are represented by striped circles.The SVC lavas are shown by solid red circles.
601 (b) $^{87}Sr/^{86}Sr$ and (d) $^{207}Pb/^{204}Pb$ vs. SiO_2 (wt. %) of St Lucia lavas showing the two trends
602 observed. SiO_2 and Sr-Nd-Pb isotopic compositions are provided in the Table 1. Tonga,
603 South Sandwich, Mariana and Aleutian compositions from Georock database:
604 http://georoc.mpch-mainz.gwdg.de/georoc/. MORB field is mid-Atlantic Ridge between
605 30°N and 30°S (data from PETDB: http://petdb.org/science.jsp/) and the Lesser Antilles
606 arc field represents data from other islands: Grenada from Thirlwall and Graham (1984);
607 Soufrière, St Vincent from Heath et al. (1998); Dominica and Martinique from Davidson
608 (1986, 1987) and Davidson and Wilson (2011); Mt Misery, St Kitts from Toothill et al.
609 (2007), The Quill, Statia from Davidson and Wilson (2011); Saba from Sherman (1992).
610

611 Figure 3: $^{87}Sr/^{86}Sr$ isotope ratios of twenty plagioclase phenocrysts showing isotopic
612 disequilibrium among four of the most contaminated SVC lavas. The whole rock value for

each lava is shown as a horizontal line. For all data, 2 standard errors (SE) are < 0.000014 which is smaller than the symbol size. Data are presented in Table 2.

Figure 4: $\delta^{18}O$ values of phenocrysts from SVC and Pre-SVC lavas (symbols as in Fig.2). Plag = plagioclase; Qz = quartz, Py = pyroxene; Am = amphibole and MA = mantle range. Mantle ranges are from Bindeman and Valley (2002) for Quartz (from differentiation of mantle derived magma), Chazot et al. (1997) for pyroxene and amphibole and Eiler et al. (2000) for plagioclase (defined using crystals from samples with SiO_2 similar to Pre-SVC mafic lavas). 2 sd reproducibility error of the O isotope technique was typically ± 0.4 ‰ (see section 3.3).

Figure 5: Correlation between mineral $\delta^{18}O$ and whole rock $^{87}Sr/^{86}Sr$ and SiO_2 (symbols as in Fig. 2). (a,b) plagioclase and pyroxene $\delta^{18}O$ vs. whole rock $^{87}Sr/^{86}Sr$. (c,d) plagioclase and pyroxene $\delta^{18}O$ vs. whole rock SiO_2. Mixing models between mantle ($\delta^{18}O$ = +5.7 ‰) and sediment with $\delta^{18}O$ of +20 ‰ (1) and +35 ‰ (2) are also shown. Oxygen concentration is assumed to be the same for both the mantle and the sediment in the model. MA = MORB mantle range with $\delta^{18}O$ from Eiler et al. (2000) and $^{87}Sr/^{86}Sr$ from mid-Atlantic Ridge between 30°N and 30°S (data from PETDB: http://petdb.org/science.jsp/). In the schematic inset $[Sr]_C$ is the Sr concentration in sediment and $[Sr]_M$ is the Sr concentration in the mantle during source contamination (SC) and in the melt during crustal assimilation (CA).

Figure 6: Variation of SiO_2 and CaO with MgO composition with the Pre-SVC and SVC lavas. Upper continental crust composition is from Rudnick and Fountain (1995). The schematic inset illustrates the impact of incorporation of large amounts of terrigenous

sediment into the mantle source on primitive magma compositions. The same concept can be applied to CaO versus MgO, for large amounts of carbonate rich sediment.

Figure 7: Schematic model for the St Lucia magmatic plumbing system through time to explain the Pre-SVC and SVC whole rock and mineral isotopic compositions. The bottom panel illustrates the storage, intrusion and eruption of the Pre-SVC lavas. The magma storage is limited and/or occurs mostly in the mantle and in the oceanic crust: assimilation of sediment (purple) is limited. The middle panel illustrates the development of a large SVC andesitic complex at shallower depth where active assimilation of sediment takes place. Finally, the upper panel illustrates the evolution of the SVC dacite from the andesite. Minimal or no assimilation occurs at this stage.

Fig. 1

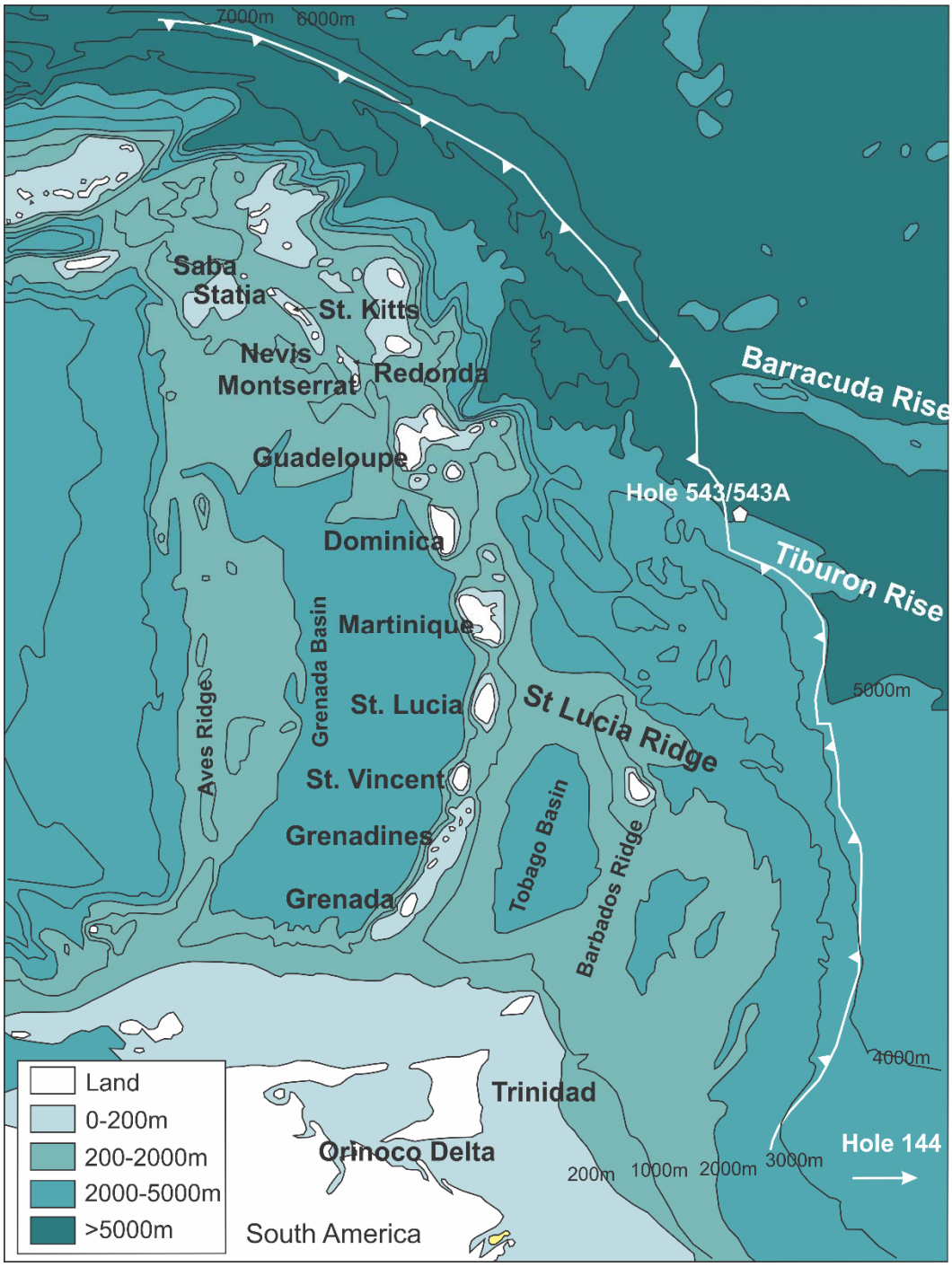
7000m
6000m
Saba
Statia
St. Kitts
Nevis
Montserrat
Redonda
Guadeloupe
Dominica
Martinique
St. Lucia
St. Vincent
Grenadines
Grenada
Aves Ridge
Grenada Basin
Barracuda Rise
Hole 543/543A
Tiburon Rise
5000m
St Lucia Ridge
Tobago Basin
Barbados Ridge
4000m
Trinidad
Orinoco Delta
South America
200m
1000m
2000m
3000m
Hole 144
Land
0-200m
200-2000m
2000-5000m
>5000m

Fig. 2

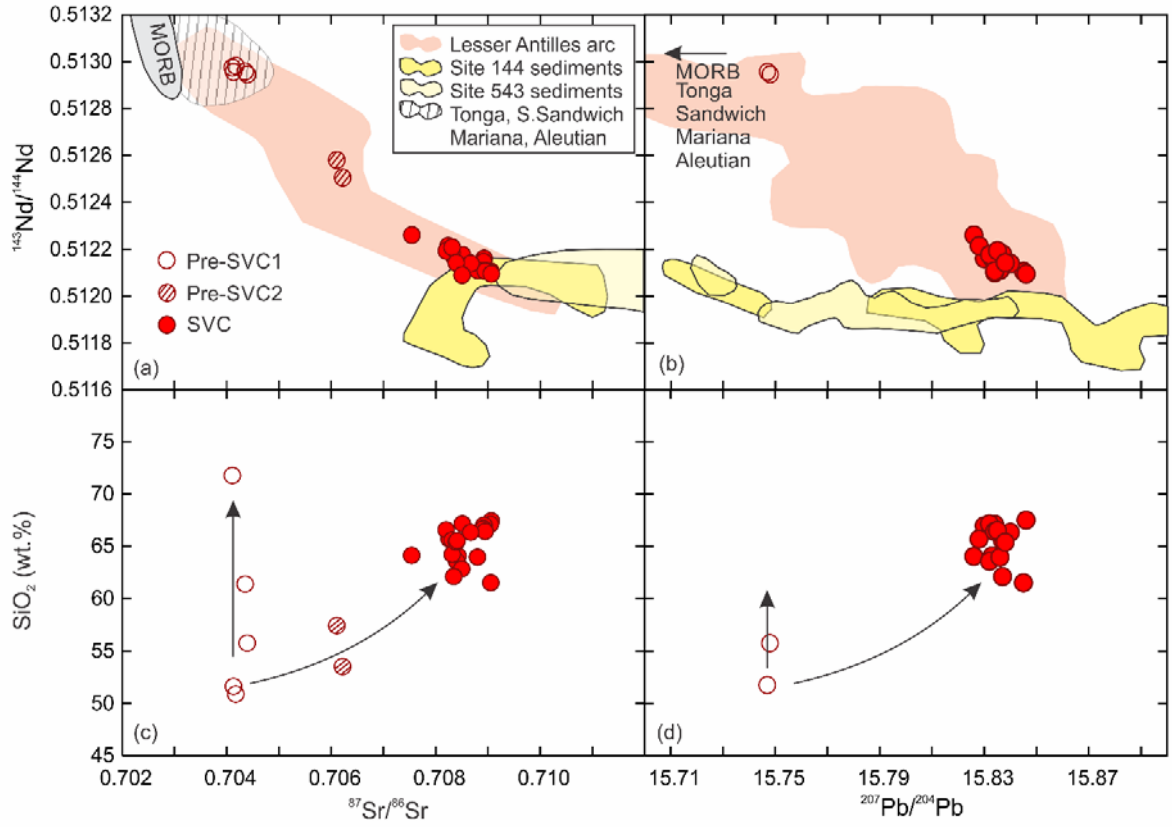
MORB
Lesser Antilles arc
Site 144 sediments
Site 543 sediments
Tonga, S.Sandwich
Mariana, Aleutian
Pre-SVC1
Pre-SVC2
SVC
(a)
(b)
MORB
Tonga
Sandwich
Mariana
Aleutian
$^{143}Nd/^{144}Nd$
0.5132
0.5130
0.5128
0.5126
0.5124
0.5122
0.5120
0.5118
0.5116
SiO_2 (wt.%)
75
70
65
60
55
50
45
(c)
(d)
0.702
0.704
0.706
0.708
0.710
$^{87}Sr/^{86}Sr$
15.71
15.75
15.79
15.83
15.87
$^{207}Pb/^{204}Pb$

Fig. 3

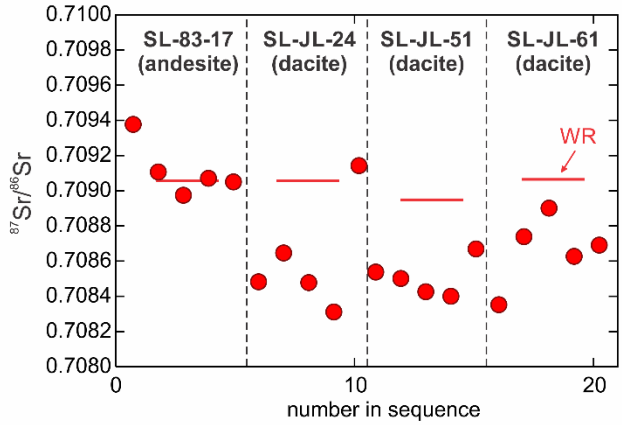
SL-83-17
(andesite)
SL-JL-24
(dacite)
SL-JL-51
(dacite)
SL-JL-61
(dacite)
WR
$^{87}Sr/^{86}Sr$
0.7100
0.7098
0.7096
0.7094
0.7092
0.7090
0.7088
0.7086
0.7084
0.7082
0.7080
0
10
20
number in sequence

Fig. 4

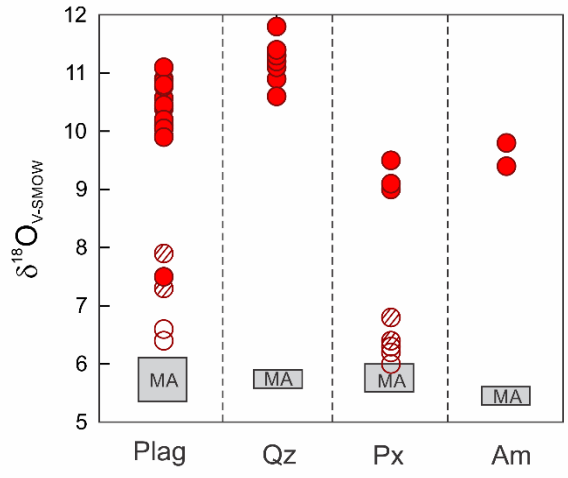
$\delta^{18}O_{V\text{-}SMOW}$
12
11
10
9
8
7
6
5
MA
MA
MA
MA
Plag
Qz
Px
Am

Fig. 5

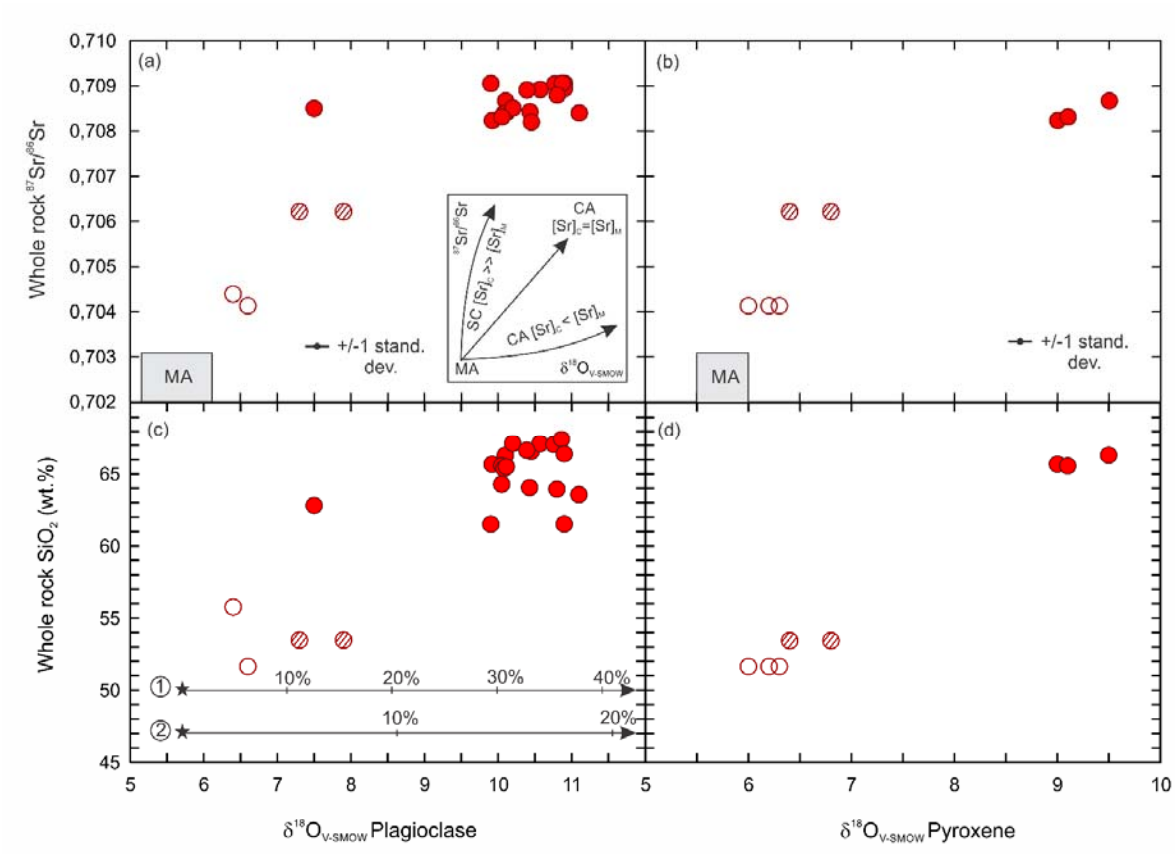
(a)
(b)
(c)
(d)
Whole rock $^{87}Sr/^{86}Sr$
Whole rock SiO_2 (wt.%)
$\delta^{18}O_{V\text{-}SMOW}$ Plagioclase
$\delta^{18}O_{V\text{-}SMOW}$ Pyroxene
MA
+/-1 stand. dev.
$^{87}Sr/^{86}Sr$
SC $[Sr]_c >> [Sr]_\mu$
CA $[Sr]_c = [Sr]_\mu$
CA $[Sr]_c < [Sr]_\mu$
$\delta^{18}O_{V\text{-}SMOW}$
10%
20%
30%
40%
①
②

Fig. 6

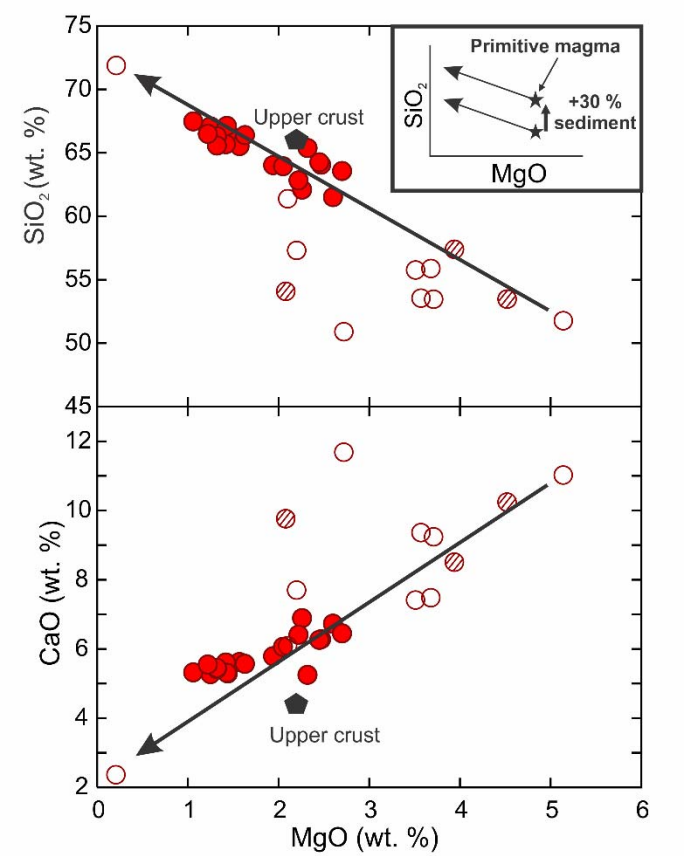
Primitive magma
+30 %
sediment
SiO2
MgO
Upper crust
SiO2 (wt. %)
CaO (wt. %)
Upper crust
MgO (wt. %)

Fig. 7

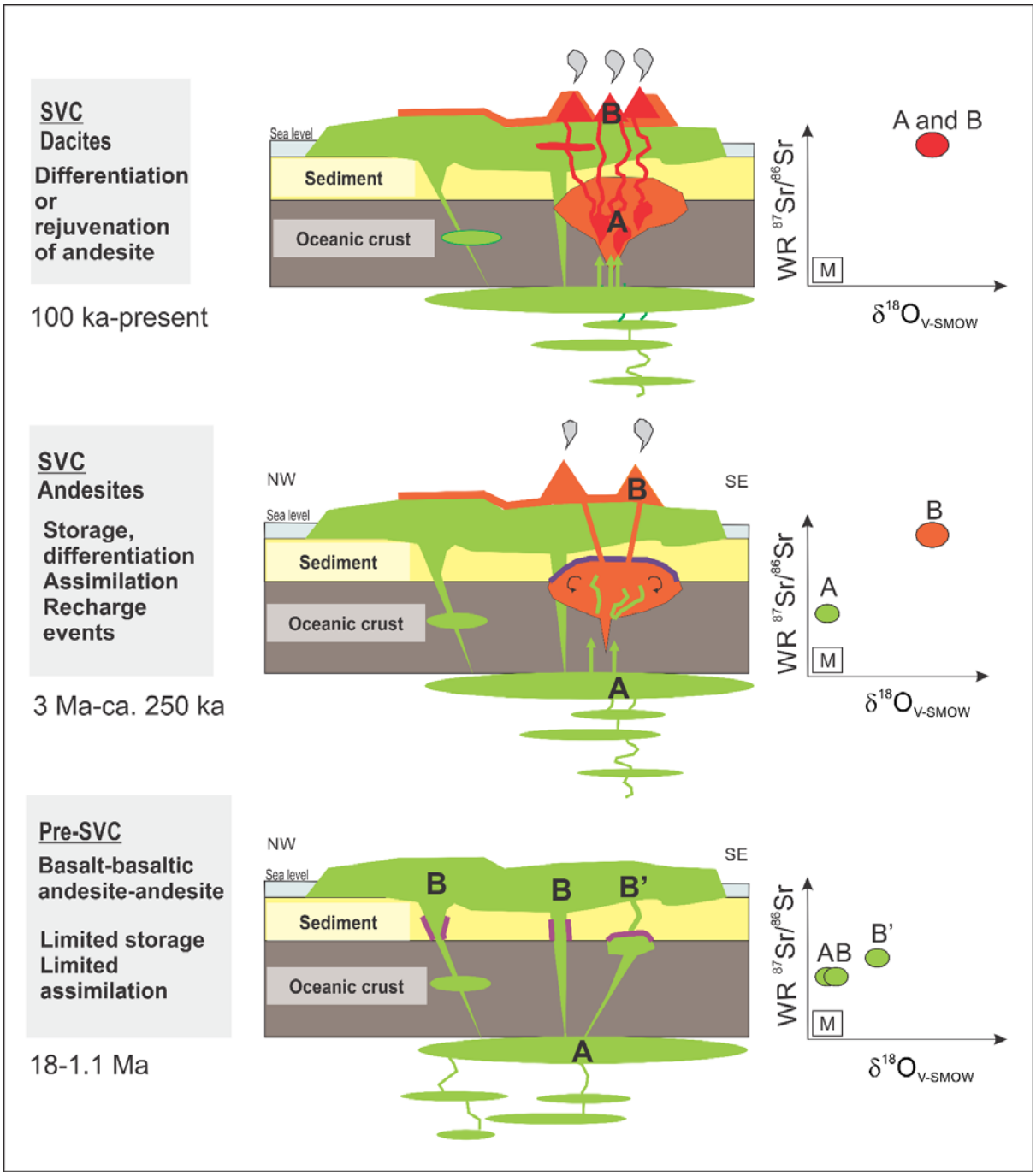
SVC
Dacites
Differentiation or rejuvenation of andesite
100 ka-present
Sea level
Sediment
Oceanic crust
B
A
A and B
WR $^{87}Sr/^{86}Sr$
M
$\delta^{18}O_{V\text{-}SMOW}$
SVC
Andesites
Storage, differentiation
Assimilation
Recharge events
3 Ma-ca. 250 ka
NW
SE
Sea level
Sediment
Oceanic crust
B
A
WR $^{87}Sr/^{86}Sr$
M
$\delta^{18}O_{V\text{-}SMOW}$
Pre-SVC
Basalt-basaltic andesite-andesite
Limited storage
Limited assimilation
18-1.1 Ma
NW
SE
Sea level
Sediment
Oceanic crust
B
B
B'
A
AB
B'
WR $^{87}Sr/^{86}Sr$
M
$\delta^{18}O_{V\text{-}SMOW}$

Table 1: St Lucia whole rock Sr-Nd-Pb isotopic compositions and the corresponding SiO_2 and CaO content in wt.%. SiO_2 and CaO concentrations are from Davidson (1987) and Lindsay et al. (2013), and normalised to 100% volatile free.

Durham University							
Sample name	**$^{143}Nd/^{144}Nd$**	**$^{87}Sr/^{86}Sr$**	**$^{206}Pb/^{204}Pb$**	**$^{207}Pb/^{204}Pb$**	**$^{208}Pb/^{204}Pb$**	**SiO_2**	**CaO**
SL8303	0.512261	0.707542	19.728	15.826	39.438	64	5.79
SL8308	0.512161	0.708925	19.721	15.830	39.457	66.9	5.36
SL8312	0.512147	0.708914	-	-	-	66.7	5.29
SL8315	0.512169	0.708346	19.760	15.837	39.467	62.1	6.89
SL8316	0.512161	0.70843	19.737	15.833	39.466	64.1	6.28
SL8317	0.512106	0.709056	19.797	15.845	39.489	61.5	6.73
SL8319	0.51218	0.708412	19.748	15.837	39.479	65.5	5.62
SL8324	0.512215	0.708237	19.730	15.828	39.446	65.7	5.61
SL8325	0.512946	0.704394	19.291	15.748	38.930	55.8	7.41
SL8344	0.512957	0.704132	19.341	15.747	39.110	51.8	11.02
SL-JL-22	0.512178	0.708402	19.753	15.832	39.471	63.6	6.45
SL-JL-23	0.512111	0.708801	19.779	15.836	39.479	63.9	6.06
SL-JL-24	0.512101	0.709049	19.770	15.834	39.484	67.1	5.27
SL-JL-33	0.512177	0.708511	19.759	15.832	39.473	67.1	5.31
SL-JL-51	0.512108	0.708946	19.766	15.834	39.484	66.4	5.57
SL-JL-57	0.51214	0.708672	19.773	15.840	39.489	66.3	5.42
SL-JL-61	0.512096	0.709063	19.782	15.846	39.528	67.5	5.32
SL-JL-79	0.512187	0.708313	-	-	-	65.6	5.45
SL-JL-83	0.512194	0.708202	19.758	15.835	39.483	66.5	5.55
SL-JL-84	0.512209	0.708313	-	-	-	64.3	6.26
Macquarie University							
Sample name	**$^{143}Nd/^{144}Nd$**	**$^{87}Sr/^{86}Sr$**	**$^{206}Pb/^{204}Pb$**	**$^{207}Pb/^{204}Pb$**	**$^{208}Pb/^{204}Pb$**	**SiO_2**	**CaO**
SL8326	0.512505	0.706219	-	-	-	53.5	10.24
SL8339	0.51258	0.706106	-	-	-	57.4	8.53
SL8341	0.512958	0.704354	-	-	-	61.4	6.09
SL8342	0.512983	0.704174	-	-	-	50.9	11.68
SL8345	0.512975	0.704109	-	-	-	71.9	2.37
SL-JL-1	0.512143	0.708393	19.757	15.838	39.489	65.4	5.25
SL-JL-2	0.512091	0.708504	-	-	-	62.8	6.41

Table 2: $^{87}Sr/^{86}Sr$ of single plagioclase grains separated from four of the most continental-like SVC hand samples. Whole rock SiO_2 (wt. %) and $^{87}Sr/^{86}Sr$ are also shown (see Table 1 for references).

Sample name	$^{87}Sr/^{86}Sr$	2 SE	Whole rock $^{87}Sr/^{86}Sr$	Whole rock SiO_2
SL8317Pl1	0.709377	0.000010		
SL8317Pl2	0.709107	0.000009		
SL8317Pl3	0.708974	0.000013	0.709056	61.5
SL8317Pl4	0.709071	0.000009		
SL8317Pl5	0.709050	0.000008		
SL-JL-24Pl6	0.708482	0.000006		
SL-JL-24Pl7	0.708646	0.000005		
SL-JL-24Pl8	0.708478	0.000010	0.709049	67.1
SL-JL-24Pl9	0.708311	0.000007		
SL-JL-24Pl10	0.709143	0.000009		
SL-JL-51Pl11	0.708538	0.000007		
SL-JL-51Pl12	0.708501	0.000010		
SL-JL-51Pl13	0.708426	0.000007	0.708946	66.4
SL-JL-51Pl14	0.708400	0.000007		
SL-JL-51Pl15	0.708669	0.000005		
SL-JL-61Pl16	0.708352	0.000008		
SL-JL-61Pl17	0.708739	0.000006		
SL-JL-61Pl18	0.708901	0.000007	0.709063	67.5
SL-JL-61Pl19	0.708627	0.000007		
SL-JL-61Pl20	0.708691	0.000006		

Table 3: $\delta^{18}O$ of single mineral separates from St Lucia along with the corresponding whole rock SiO_2 (wt.%) and Sr isotopic composition. WR = whole rock. SiO_2 content from Davidson (1987) and Lindsay et al. (2013) and normalised to 100% volatile free.

Sample name	Kind of mineral	Group	$\delta^{18}O_{V\text{-}SMOW}$ (‰)	WR SiO_2	WR $^{87}Sr/^{86}Sr$
8308Pl	Plagioclase	SVC	10.6	66.9	0.708925
8312Pl	Plagioclase	SVC	10.4	66.7	0.708914
8316Pl	Plagioclase	SVC	10.4	64.1	0.70843
8317Pl	Plagioclase	SVC	10.9	61.5	0.709056
8317Pl	Plagioclase	SVC	9.9	61.5	0.709056
8319Pl	Plagioclase	SVC	10.1	65.5	0.708412
8324Pl	Plagioclase	SVC	9.9	65.7	0.708237
8325Pl	Plagioclase	Pre-SVC1	6.4	55.8	0.704394
8326Pl1	Plagioclase	Pre-SVC2	7.3	53.5	0.706219
8326Pl2	Plagioclase	Pre-SVC2	7.9	53.5	0.706219
8344Pl	Plagioclase	Pre-SVC1	6.6	51.8	0.704132
JL01Pl	Plagioclase	SVC	10.1	65.4	0.708393
JL02Pl	Plagioclase	SVC	7.5	62.8	0.708504
JL22Pl	Plagioclase	SVC	11.1	63.6	0.708402
JL23Pl	Plagioclase	SVC	10.8	63.9	0.708801
JL24Pl	Plagioclase	SVC	10.8	67.1	0.709049
JL33Pl	Plagioclase	SVC	10.2	67.1	0.708511
JL51Pl	Plagioclase	SVC	10.9	66.4	0.708946
JL57Pl	Plagioclase	SVC	10.1	66.3	0.708672
JL61Pl	Plagioclase	SVC	10.9	67.5	0.709063
JL83Pl	Plagioclase	SVC	10.5	66.5	0.708202
JL84Pl	Plagioclase	SVC	10.0	64.3	0.708313
JL79Pl	Plagioclase	SVC	10.0	65.6	0.708313
8303Qz	Quartz	SVC	11.2	64.0	0.707542
JL01Qz	Quartz	SVC	11.3	65.4	0.708393
JL02Qz	Quartz	SVC	11.4	62.8	0.708504
JL22Qz	Quartz	SVC	11.3	63.6	0.708402
JL23Qz	Quartz	SVC	10.9	63.9	0.708801
JL24Qz	Quartz	SVC	11.4	67.1	0.709049
JL33Qz	Quartz	SVC	11.1	67.1	0.708511
JL51Qz	Quartz	SVC	11.3	66.4	0.708946
JL57Qz	Quartz	SVC	10.6	66.3	0.708672
JL61Qz	Quartz	SVC	11.8	67.5	0.709063
8324Opx	Orthopyroxene	SVC	9	65.7	0.708237
8326Py1	Clinopyroxene	Pre-SVC2	6.8	53.5	0.706219
8326Py2	Clinopyroxene	Pre-SVC2	6.4	53.5	0.706219
8344Py1	Clinopyroxene	Pre-SVC1	6.0	51.8	0.704132
8344Py2	Clinopyroxene	Pre-SVC1	6.2	51.8	0.704132
8344Py3	Clinopyroxene	Pre-SVC1	6.3	51.8	0.704132
JL57Opx	Orthopyroxene	SVC	9.5	66.3	0.708672
JL79Opx	Orthopyroxene	SVC	9.1	65.6	0.708313
8308Am	Amphibole	SVC	9.8	66.9	0.708925
JL33Am	Amphibole	SVC	9.4	67.1	0.708511